Theory of Ferromagnetism in Diluted Magnetic Semiconductor Quantum Wells

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We present a mean field theory of ferromagnetism in diluted magnetic semiconductor quantum wells. When subband mixing due to exchange interactions between quantum well free carriers and magnetic impurities is neglected, analytic result can be obtained for the dependence of the critical temperature and the spontaneous magnetization on the distribution of magnetic impurities and the quantum well width. The validity of this approximate theory has been tested by comparing its predictions with those from numerical self-consistent field calculations. Interactions among free carriers, accounted for using the local-spin-density approximation, substantially enhance the critical temperature. We demonstrate that an external bias potential can tune the critical temperature through a wide range.

The discovery of carrier-mediated ferromagnetism[1–4] in diluted magnetic semiconductors (DMS) has opened a broad and relatively unexplored frontier for both basic and applied research. The interplay between collective magnetic properties and semiconductor transport properties in these systems presents a rich phenomenology and offers the prospect of new functionality in electronic devices. The possibilities for manipulating this interplay are especially varied when the free carrier system is quasi-two-dimensional[5–7] because of the dependence of system properties on the subband wavefunction. In this article we address the dependence of the ferromagnetic critical temperature T_c of two-dimensional DMS ferromagnets on the spatial distribution of magnetic ions, the subband wavefunction of the free carrier system, and on their interplay. Our analysis is based on a formulation of the mean-field theory for carrier-induced ferromagnetism[8] in a DMS which was developed in earlier work[9] and is intended to be useful for any spatially inhomogeneous system. We find that T_c is quite sensitive to the relative distributions of electrons and magnetic ions and can be altered *in situ* by applying a gate voltage.

For definiteness we address the case of a $\langle 001 \rangle$ growth direction 2D hole gas in the $Ga_{1-x}Mn_xAs$ DMS system. Much of our analysis, however, applies equally well to other interfaces and to other DMS systems with cubic host semiconductors and, with one important caveat which we mention below, also to the case of a 2D electron gas mediated ferromagnetism. Our theory is based on an envelope function description of the valence band electrons and a spin representation of their kinetic-exchange interaction[10] with d-electrons[11] on the $S = 5/2$ Mn^{++} ions:

$$\mathcal{H} = \mathcal{H}_m + \mathcal{H}_f + J_{pd} \sum_{i,I} \vec{S}_I \cdot \vec{s}_i \delta(\vec{r}_i - \vec{R}_I), \qquad (1)$$

where i labels a free carrier, I labels a magnetic ion and the exchange interaction energy $J_{pd} \approx 0.15$ eV nm^3. In Eq.(1) $\mathcal{H}_m$ is the Hamiltonian of the magnetic ions, $\mathcal{H}_f$ is the four-band Luttinger Hamiltonian for free carriers in the valence band, $\vec{S}$ is the magnetic ion spin and $\vec{s}$ is the electron-spin operator projected onto the $j = 3/2$ valence band manifold of the Luttinger Hamiltonian. We assume here that the 2D free carrier density is sufficiently low that only the lowest energy heavy-hole subband is occupied, and that the quantum well of interest is sufficiently narrow that mixing between light-hole and heavy-hole bands can be neglected. These simplifications lead[12] to a single parabolic band with the in-plane effective mass $m_\parallel^* \approx 0.11 m_0$ and the out-of-plane mass $m_z^* \approx 0.38 m_0$. The two spin-states in this band have definite $j_z = \pm 3/2$ and definite $s_z = \pm 1/2$; the projection of the transverse spin components onto this band is zero so that the kinetic exchange interaction has an Ising character.[13]

Our mean-field theory[9] is derived in a spin-density-functional theory framework and leads to a set of physically transparent coupled equations. In the absence of external fields, the mean polarization of a magnetic ion is given by

$$\langle m \rangle_I = S B_S\big(J_{pd} S\big[n_\uparrow(\vec{R}_I) - n_\downarrow(\vec{R}_I)\big]/2k_BT\big); \qquad (2)$$

where $B_S(x)$ is the Brillouin function,

$$B_S(x) = \frac{2S+1}{2S}\coth\Big(\frac{2S+1}{2S}x\Big) - \frac{1}{2S}\coth\Big(\frac{1}{2S}x\Big)$$
$$\approx \frac{S+1}{3S}x - \frac{(S+1)(2S^2+2S+1)}{90S^3}x^3,\ x \ll 1\,. \qquad (3)$$

The electron spin-densities $n_\sigma(\vec{r})$ are determined by solving the Schrödinger equation for electrons which experience a spin-dependent potential due to kinetic exchange with the polarized magnetic ions:

$$\Big[-\frac{1}{2m_\parallel^*}\Big(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\Big) - \frac{1}{2m_z^*}\frac{\partial^2}{\partial z^2} + v_{es}(\vec{r})$$

$$+v_{xc,\sigma}(\vec{r}) - \frac{\sigma}{2}h_{pd}(\vec{r})\Big]\psi_{k,\sigma}(\vec{r}) = \epsilon_{k,\sigma}\psi_{k,\sigma}(\vec{r}) \; ; \qquad (4)$$

$$n_\sigma(\vec{r}) = \sum_k f(\epsilon_{k,\sigma})|\psi_{k,\sigma}(\vec{r})|^2 \; . \qquad (5)$$

In these equations $v_{es}(\vec{r})$ is the electrostatic potential, including band offset and ionized impurity contributions, $v_{xc,\sigma}(\vec{r})$ is an exchange-correlation potential on which we comment further below, and $f(\epsilon)$ is the Fermi distribution function. The spin-dependent kinetic-exchange potential,

$$h_{pd}(\vec{r}) = J_{pd}\sum_I \delta(\vec{r} - \vec{R}_I)\langle m\rangle_I \qquad (6)$$

is non-zero only in the ferromagnetic state. In the following we assume that the magnetic ions are randomly distributed and dense on a scale set by the free carrier Fermi wavevector in the $\hat{x} - \hat{y}$ plane, and that their density in the growth direction, $c(z)$, can be precisely controlled.[7] This allows us to take a continuum limit where the magnetic ion polarization and the kinetic-exchange potential depends only on the growth direction coordinate and

$$h_{pd}(z) = J_{pd}\; c(z)\; \langle m\rangle(z). \qquad (7)$$

In the following we discuss how the ferromagnetic transition temperature T_c depends on $c(z)$.

The T_c calculation is greatly simplified when the spin-dependence of the exchange correlation potential is neglected and the quantum well width w is small enough to make subband mixing due to kinetic exchange interactions negligible, i.e., when $Jc(z) \ll \hbar^2/m_z^* w^2$. We see later that the theory which results from these approximations is normally quite accurate. With subband mixing neglected, the only effect of the mean-field kinetic-exchange interaction on the 2D carriers is to produce a rigid spin-splitting of the 2D bands, given by the first-order perturbation theory expression:

$$\epsilon_\downarrow - \epsilon_\uparrow = J_{pd}\int dz' \langle m\rangle(z')c(z')|\psi(z')|^2 \qquad (8)$$

where $\psi(z)$ is the growth direction envelope function of the lowest subband. For the case of a quantum well with infinite barriers, $\psi(z) = \sqrt{2/w}\cos(\pi z/w)$. The electron spin-polarization can then be obtained by summing over occupied free-carrier states. It follows that, as long as both spin-$\uparrow$ and spin-$\downarrow$ bands are partly occupied,

$$n_\uparrow(z) - n_\downarrow(z) = \frac{m_\parallel^*}{2\pi\hbar^2}|\psi(z)|^2 J_{pd}\int dz' \langle m\rangle(z')c(z')|\psi(z')|^2 \; . \qquad (9)$$

Eq. (9) can be used to eliminate the free-carrier spin-polarization from Eq. (2) and to obtain a self-consistent equation for the function $\langle m\rangle(z)$. The equation depends only on the kinetic-exchange coupling constant J_{pd}, the free-carrier in-plane effective mass $m_\parallel^*$, the subband wavefunction $\psi(z)$, and the magnetic ion distribution function $c(z)$. It is important to realize that the entire system behaves collectively. It has a single critical temperature which depends on the function $c(z)$, rather than a z-dependent T_c like that which appears in a naive adaptations of the bulk RKKY theory for T_c to quasi-2D systems.

An explicit expression for the critical temperature can be obtained by linearizing the self-consistent equation (2). Expanding the Brillouin function to the first order in its argument gives the homogeneous linear integral equation

$$\langle m\rangle(z) = \frac{1}{k_B T}\int dz' K(z,z')\langle m\rangle(z') \qquad (10)$$

where the kernel

$$K(z,z') = \frac{S(S+1)J_{pd}^2}{12}\frac{m_\parallel^*}{\pi\hbar^2}|\psi(z)|^2 c(z')|\psi(z')|^2. \qquad (11)$$

Since the linearized equation is satisfied only at the critical temperature, $k_B T_c$ is equal to the largest eigenvalue of $K(z,z')$, the eigenvalue corresponding to eigenfunction $f(z) \propto |\psi(z)|^2$. It follows that

$$T_c = \frac{S(S+1)}{12}\frac{J_{pd}^2}{k_B}\frac{m_\parallel^*}{\pi\hbar^2}\int dz|\psi(z)|^4 c(z) \; . \qquad (12)$$

The critical temperature is proportional to J_{pd}^2 and $m_\parallel^*$, is independent of 2D carrier density, and for a system with uniformly distributed magnetic impurities is inversely proportional to the quantum well width, as illustrated by the solid line in Fig. 1. The accuracy of Eq. (12) has been tested by numerically solving the set of self-consistent Eqs. (2)-(7) together with the Poisson equation for $v_{es}(z)$ and the local-spin-density-approximation (LSDA) formula[14] for $v_{xc,\sigma}(z)$. If exchange and correlation effects are neglected ($v_{xc,\sigma}(z) = 0$), the self-consistent numerical results represented by circles in Fig. 1 are identical to those obtained from Eq. (12) for quantum well widths $w = 10$, 15, and 20 nm. Interactions among the 2D carriers substantially enhance the critical temperature and also cause a strong dependence of T_c on the 2D carrier density N, as illustrated by squares ($N = 1\times10^{11}$ cm^2) and triangles ($N = 0.5\times10^{11}$ cm^2) in Fig. 1. Note that unlike the 3D case, where T_c is proportional to Fermi wavevector in the non-interacting limit[8,9] and is an increasing function of density even if the interactions are included,[9] the critical temperature for all three quantum wells of Fig. 1 is larger at lower density, $N = 0.5\times10^{11}$ cm^2, where the interactions are stronger.[15]

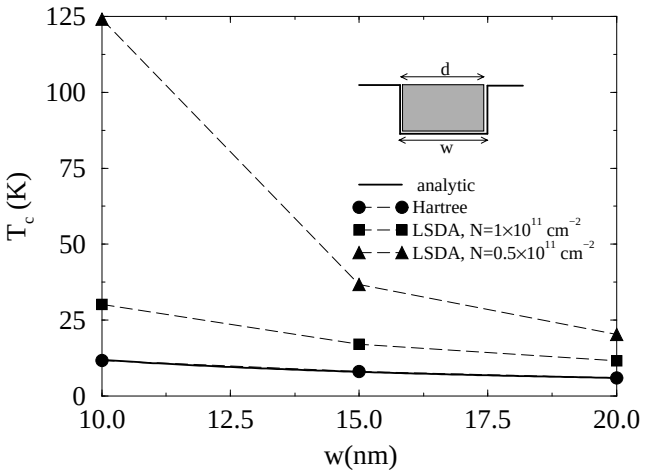

FIG. 1. Ferromagnetic critical temperature of a quasi-2D DMS as a function of quantum well width w for fixed 3D magnetic ion density $c = 1\text{nm}^{-3}$; inset: schematic diagram of the quantum well with the shaded portion of width d doped with magnetic ions. In this figure $d = w$. The non-interacting free carrier results were calculated both by solving the full self-consistent equations (circles) and by using the approximate T_c expression derived in the text (solid line), confirming the accuracy of the latter approach. Squares and triangles represent the full numerical LSDA calculations for 2D carrier densities $N = 1 \times 10^{11}$ cm^2 and $N = 0.5 \times 10^{11}$ cm^2, respectively.

Before discussing the implications of the expression (12) for T_c we comment on its relation to the RKKY theory[8] of carrier induced ferromagnetism in DMS's. As applied to bulk 3D electron systems the two approaches are fundamentally equivalent, although there are differences in detail which can be important. The principle difference is that the RKKY theory treats the free-carrier magnetic-ion interaction perturbatively. As a result both approaches give the same value for the critical temperature where the magnetization vanishes; differences appear at lower temperatures where the electron system is strongly or completely spin-polarized and a perturbative treatment of its interactions with the magnetic ions fails. Both approaches fail to account for the retarded character of the free-carrier mediated interaction between magnetic ions in these low density systems, and for correlated quantum fluctuations in magnetic ion and free carrier subsystems which are important for some properties.[16] Both approaches are able to account for interaction effects in the free carrier system, which increase the tendency toward ferromagnetism as illustrated in Fig. 1, and in the density-functional approach appear in the spin-dependence of the exchange correlation potential. It is in inhomogeneous cases, such as the reduced dimension situation considered here, that the density functional approach has an advantage. Our approach provides a simple description of the collective behavior of the quasi-2D ferromagnet and is able to handle geometric features which can be engineered to produce desired properties. For example we show in Fig. 2 a plot of the dependence of the critical temperature on the fraction d of the quantum well occupied by magnetic ions. When the integrated 2D density of magnetic ions ($c\ d$) is fixed, the critical temperature increases as magnetic impurities are transferred toward the center of the quantum well where the free-carrier wavefunction has its maximum. For fixed 3D density c the number of magnetic impurities decreases with decreasing d. This effect is stronger than the increase of the overlap between the carrier wavefunction and magnetic ion distribution resulting in the decrease of T_c, as shown in the inset of Fig. 2. In both cases the analytic results of Eq. (12) are in qualitative agreement with the full numerical LSDA calculations.

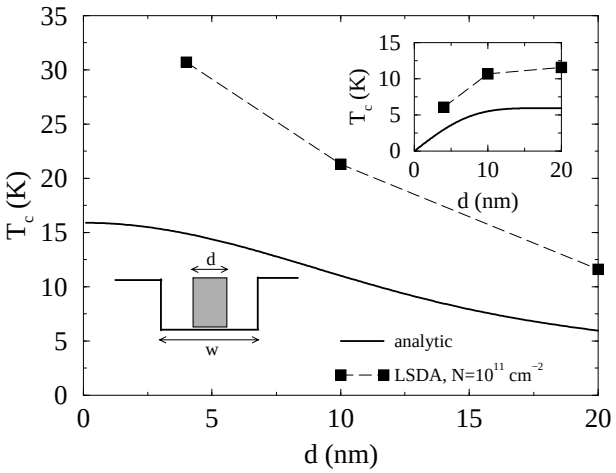

FIG. 2. Dependence of T_c on the central portion d of the quantum well ($w = 20$ nm) occupied by magnetic ions. The main graph is for fixed 2D magnetic ion density, $c\ d = 20\ \text{nm}^{-2}$, while the inset is for the case of fixed 3D magnetic ion density, $c = 1\ \text{nm}^{-3}$.

One remarkable feature of quasi-2D ferromagnetic DMS's is the possibility of tuning T_c through a wide range in situ, by the application of a gate voltage. In Fig. 3 we illustrate this for the case of a $w = 10$ nm quantum well with magnetic ions covering only a $d = 3$ nm portion near one edge. Even for such a narrow quantum well, the critical temperature can be varied over an order of magnitude by applying a bias voltage which draws electrons into the magnetic ion region.

In closing we remark that for several reasons DMS ferromagnetism mediated by 2D electron systems is likely to occur only at inaccessibly low temperatures. One important factor is the smaller value of the exchange interaction parameter in the conduction band case. For example for the typical n-type II-VI DMS quantum well[3], $(\text{Zn}_{1-x}\text{Cd}_x\text{Se})_{m-f}(\text{MnSe})_f$,[7] the $s-d$ exchange $J_{sd} \sim 4 \times 10^{-3}$ eV nm^3. Our theory then gives $T_c \sim 10$ mK for a 10 nm wide quantum well. Equally important, however, is the weak magnetic anisotropy expected in the conduction band case, which will lead to soft spin-wave

collective modes[17] and substantial T_c suppression.

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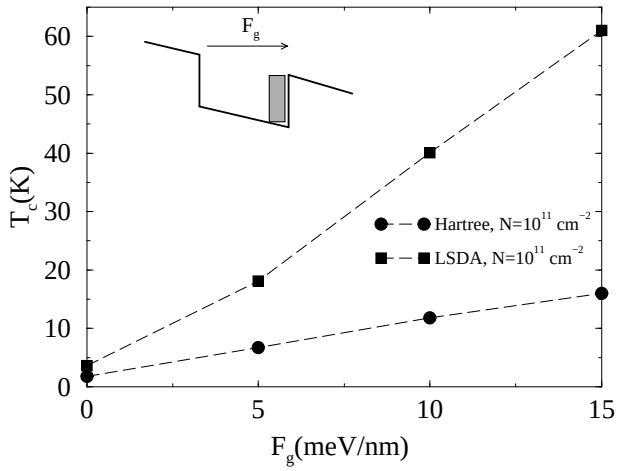

FIG. 3. Dependence of T_c on bias voltage applied across the $w = 10$ nm wide quantum well partially occupied by magnetic ions over a distance of 3 nm near one edge of the quantum well. Circles (squares) represent results of the full numerical self-consistent calculations without (with) the exchange-correlation potential.

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[11] We assume free carrier screening makes the central cell corrections required for isolated Mn acceptors in GaAs unimportant. See A.K. Bhattacharjee and C. Benoit á la Guillaume, Solid State Commun. **113**, 17 (2000) and work cited therein.

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[13] More generally there is a delicate interplay between size-quantization controlled heavy-light subband mixing and kinetic-exchange interaction anisotropy which can be accounted for if the geometry of a particular sample is accurately known.

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